Health Consultation

Lawrence Livermore National Laboratory
Big Trees Park 1998 Sampling
Livermore, California

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Federal Facilities Assessment Branch
Division of Health Assessment and Consultation
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BACKGROUND

In April 1999, the California Department of Health Services, Environmental Health Investigations Branch (CDHS-EHIB) under a cooperative agreement with the Agency for Toxic Substances and Disease Registry (ATSDR) released the consultation "Lawrence Livermore National Laboratory, Plutonium Contamination in Big Trees Park" [1]. In that public health consultation, an analysis of the plutonium-associated radiological dose that might be received by individuals using the park was determined not to be a public health concern. Nonetheless, the main question not answered in that document was how the plutonium (Pu[1]) reached the park. ATSDR and CDHS-EHIB made three specific recommendations regarding the need for additional sampling in Big Trees Park. These recommendations as published were as follows:

1. We recommend further sampling of Big Trees Park and the Arroyo Seco Creek sediments to determine the vertical and horizontal extent of the Pu 239 contamination.

2. We recommend further evaluation of the distribution of contaminated sludge throughout the Livermore Valley, and other areas. This would include assessing the different avenues for gathering information on where sludge may have been distributed, and if locations are identified that may have received contaminated sludge, assessing the feasibility of various approaches to characterize the potential plutonium in those areas.

3. We recommend that potential contaminant releases from sewer line ruptures be investigated in future health consultations focusing on sediments in the Livermore Valley or LLNL discharges to the sanitary sewer system.

PURPOSE OF THIS PUBLIC HEALTH CONSULTATION

ATSDR is preparing this public health consultation to address the following issues: 1) Overall quality of the data collected during the 1998 Big Trees Park sampling event; 2) evaluate the possible pathways by which Pu reached the park and; 3) if additional consideration should be given for an area-wide sampling for Pu in residential soils.

[1]Elements have been assigned either one or two letter abbreviations. The abbreviation for plutonium is Pu. When a number is associated with the name or the abbreviation, this indicates the mass of the particular element. For example, Pu 239 represents plutonium with a mass of 239. If two numbers are given such as Pu 239/340, this indicates that typical radiochemistry laboratory analyses cannot differentiate between Pu 239 and Pu 240 because the energies of their decay is almost identical and cannot be differentiated by the counting equipment.

STATEMENT OF ISSUES

A series of public meetings were held in the city of Livermore in 1997 and 1998. One outcome of these meetings was that the regulatory agencies composed of the U.S. Environmental Protection Agency (EPA), the U.S. Department of Energy (DOE) and CDHS-Radiological Health Branch (CDHS-RHB), agreed that additional sampling in Big Trees Park was warranted. DOE and the Lawrence Livermore National Laboratory (LLNL), with regulatory oversight, developed a new sampling plan. This sampling plan released for public comment, was finalized in October 1998 [2]. Included in this sampling effort was a split sampling plan in which approximately 10% of the samples collected would be supplied to ATSDR for independent analysis [3].

As stated in the sampling plan the purpose of the 1998 round of sampling was to describe methodologies, analytical procedures, and related tasks to determine how the plutonium identified in the previous public health consultation could have reached the park; that is, to collect environmental samples for a pathway analysis. The sampling plan required collection of soil samples at various locations and depths within the Big Trees Park. The major locations included grid samples to determine if the pathway was air dispersion, the current Arroyo Seco channel and the old channel before the realignment of the creek to determine if the pathway was releases of Pu from LLNL via sewer line breaks or via overland flow from the southeast quadrant of LLNL, and sampling in and around ornamental trees to determine if the pathway was use of contaminated sewage sludge as a soil amendment. Other sampling areas included a disked area done at the request of a community member to evaluate the dust and dirt as a possible inhalation and ingestion pathway, the Big Trees Park extension, and sampling in the playground of the Arroyo Seco Elementary School. A detailed discussion of the rationale associated with these locations is beyond the scope of this public health consultation but is available in the 1998 Sampling Plan document [2].

Previous Sampling Events

The first publically reported sampling of Big Trees Park was released in 1994 when the EPA National Air and Radiation Environmental Laboratory was doing confirmatory surveys of Pu contamination in the southeast quadrant of LLNL [4]. Big Trees Park and two additional city parks in Livermore were chosen to represent areas considered background. Based on these results, Pu might be elevated in the surface soils of this park and the two additional parks in Livermore. In response to the discovery of Pu in Big Trees Park, LLNL in 1995 repeated the Big Trees Park sampling for verification purposes [5]. Details of these sampling events can be found in the CDHS-EHIB and ATSDR public health consultation [1] and the referenced documents [4,5].

Because each of the previous sampling efforts had different data quality objectives, the data between these sets should not have been analyzed as a combined group for several reasons. In ATSDR's opinion, these included, but are not limited to, a lack of available inter-laboratory quality control and assurance from independent laboratories, variations in the methodologies used

for the analyses, lower limit of detection issues, and precision and accuracy for the intercomparison of data sets. These factors have been discussed at several public meetings; CDHS-EHIB, ATSDR, and the regulatory agencies have also discussed these issues through a series of correspondence. Additional discussion of the previous data sets is not warranted and, therefore, is beyond the scope of this document.

Data Quality, Quality Control and Quality Assurance Activities

For the 1998 sampling of Big Trees Park, the issues of data quality were addressed through a more in-depth oversight by the EPA and CDHS-RHB, public participation via a comment period, a split sampling plan among LLNL, EPA, and ATSDR, and regulatory analysis of the split sampling data by EPA. ATSDR involvement in this process was three-fold: 1) Serve as a central collection point for public comments; 2) Arrange for an independent laboratory analysis of approximately 10% of the collected samples and; 3) Participate in the data quality review for the purposes of this sampling event.

A data quality team was established consisting of the federal agencies, ATSDR, EPA, and DOE, and the state and laboratory entities, CDHS-RHB, and LLNL. The team was selected based on their knowledge of radiation chemistry, radiological measurement protocols, and familiarity with radiation measurement instrumentation. Identical data packages including results, quality control samples, protocols and analysis procedures were supplied and reviewed by this team. Discussions on the methods for this team review were discussed in the DOE offices in Oakland, California prior to sample collection. Present for these technical discussions were representatives from CDHS-EHIB and RHB, ATSDR, EPA, DOE, and LLNL. Additional technical deliberations and discussions of data validation issues were via conference telephone calls or via electronic mail.

The EPA Quality Assurance office in San Francisco also requested a review of the quality control and quality assurance issues by its Office of Radiation and Indoor Air, Montgomery, Alabama, and the Indoor Environments National Laboratory in Las Vegas, Nevada. The latter organization also reviewed analytical methods, quality control procedures of the participating laboratories, and evaluated the results with respect to the samples collected per the split sampling plan. The conclusion of this evaluation was that, overall, the data among the laboratories were in close agreement and "most of the data is useable without qualification" [6].

Statistical considerations for radiological data analysis

Typically, results from the analysis of radioactive samples are expressed as a 3-number set that includes the measured result, an indication of the uncertainty in the measurement, and the minimum detectable activity. Therefore, many statistical considerations must be taken into account during the evaluation of these samples, especially those samples at or near values considered background values. These considerations include method of sample collection and preservation, the methods of analysis, and laboratory quality control and assurance that the samples are handled identically. These issues all contribute to the general term "error."

Furthermore, the instruments used to measure the radioactivity also contribute to the uncertainty error, the error of the measurement denoted as the 2 sigma error (2σ). The 2σ error means that there is a 95% confidence that the reported value is correct with only a 5% chance that the "true" number is outside the range in the given interval. For example, 10 ± 3 means that the measured value is 10 with an error of 3. This is interpreted as the true number is between 7 and 13. Occasionally, the 2σ error is larger than the measured value, for example, 10 ± 15. In this situation, the sample should be considered to be a non-detect or zero.

When radiological samples with concentrations that approach either the lowest level that can be detected by the instrumentation or approach the levels associated with background, are analyzed, the 2σ error increases dramatically. Therefore, finding 2σ values approaching 50% or more of the measured value is common in these situations. Therefore, these data could be considered not to be significantly different from background or a non-detection.

For this consultation, ATSDR analyzed the data using the following procedures that were established by ATSDR prior to the start of the data evaluation:

1. Does the laboratory-derived value exceed the minimum activity or level of instrument detection (MDA)? If the value is less than the MDA, then the sample measurement is not significant and no additional evaluation is needed.

2. Does the laboratory-derived value exceed the reporting limit as outlined in the sampling plan? If the sample is above the reporting limit, continue with the evaluation.

3. Is the measured laboratory-derived measurement greater than the MDA but less than the reporting level? If so, continue with the evaluation.

4. Is the 2σ interval less than the laboratory-derived measurement and what percentage does it represent? If the 2σ is less than 25% of the laboratory reported value, consider the measurement a valid measurement. If the 2σ is more than 25% attempt to evaluate using other methods such as duplicate samples.

DISCUSSION

Overall Data Quality During the sampling, approximately 10% of the samples were sent to the Georgia Institute of Technology (Georgia Tech)[2] in Atlanta for independent analysis of Pu 238, and Pu 239/240 by alpha spectroscopic techniques, and for Americium 241 (Am 241) by low

[2]The identification of non-government affiliated private laboratories in this document is neither an endorsement nor a recommendation of their capabilities. Their mention is provided for informational purposes only. ATSDR has an Interagency Agreement with the Division of Federal Occupational Health (DFOH), Health Resources Services Administration, Department of Health and Human Services. The laboratory services of Georgia Tech were obtained through a subcontract administered by DFOH.

energy X-ray analysis with verification as needed by alpha spectroscopy. ATSDR also requested the laboratory analyze samples by gamma spectroscopy for additional products that might be present in the soil samples. Another set of samples from the same locations was supplied to LLNL for analysis by their laboratories and a third set consisting of all samples collected was supplied to General Engineering Laboratory (GEL), the prime laboratory for sample analyses, under contract to DOE. The data quality review process only allows for an intercomparison of those samples evaluated by the three laboratories. Therefore, only Pu 238, Pu 239/240, and Am 241 were considered.

Briefly, the quality of the data collected during the 1998 sampling of Big Trees Park was very good. Comparisons of the results from the three reporting laboratories were in good agreement with respect to the reported Pu 239/240 values. Therefore, ATSDR and the regulatory agencies are comfortable that the results for the remainder of the samples are a true representation of the environmental conditions in Big Trees Park. Figure 1 shows these reported values from each laboratory for the major radionuclide of concern, Pu 239/240, expressed as Pu 239. From this figure, one can see that most of the samples were indistinguishable from one laboratory to another. Furthermore, a statistical analysis these samples, called Analysis of Variance, suggest that these measured values are identical to one other.

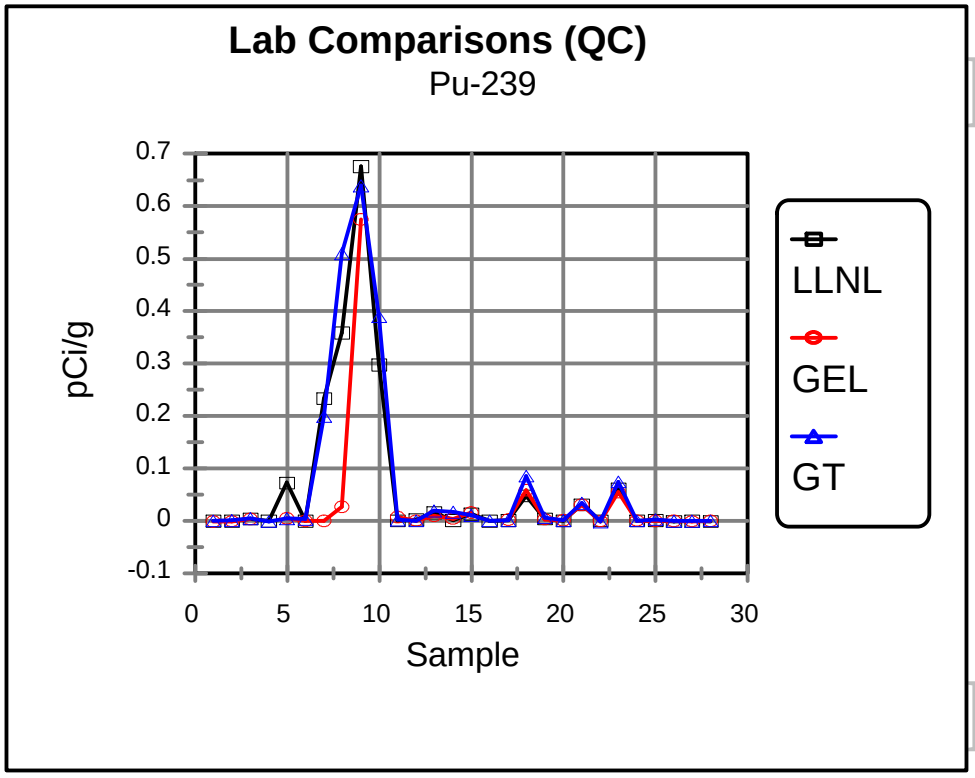

Figure 1. Interlaboratory comparison of Plutonium 239 analysis from split soil samples.

After the agencies involved with the quality assurance process were satisfied with the acceptability of the data set, the final data sets were distributed to the regulatory agencies and ATSDR. LLNL also posted the results on their World Wide Web site at the following addresses: http://www-envirinfo.llnl.gov/ or http://www-erd.llnl.gov/bigtrees/ and includes maps of the sampling locations and the sampling results. These results are used in the development of this public health consultation.

Overall, LLNL and EPA collected 379 samples for Pu 238 and Pu 239/240 analysis, 117 samples were also used for Am 241 analysis, and 101 samples were also used for metals analysis. These samples included duplicate samples as internal checks on laboratory procedures. The metals included in the analysis were those commonly found in sewage sludges - chromium, copper, lead, nickel, and zinc [2].

The sampling plan required a reporting level of Pu 239/240 at 0.005 picocuries per gram (pCi/g) equivalent to 2×10^{-4} becquerels per gram (Bq/g). This value approaches the regional background as determined by LLNL and other reports as referenced in the sampling plan [1,2,4,7]. The background values as reported by LLNL and based on approximately 200 samples, shows a typical environmental distribution of Pu 239/240 with an estimated geometric mean concentration[3] of 0.0026 pCi/g (1×10^{-4} Bq/g) [2]; Gallegos reported Pu 239/240 levels ranging from 1.2×10^{-4} Bq/g (3.2×10^{-3} pCi/g) to 2×10^{-4} Bq/g (5.4×10^{-3} pCi/g) upwind and downwind from LLNL, respectively [7]. Furthermore, converting data collected by Hardy [8] into the more common units of picocuries per gram from millicuries per square kilometer by using standard values for soil density, one estimates the Pu concentration at 2.2×10^{-4} Bq/g (6×10^{-3} pCi/g); this also agrees with LLNL data. Other estimates of background concentrations of Pu 239/240 suggests regional differences exist possibly related to weather conditions and soil disturbing activities which have been shown to alter radionuclide concentrations in soils [9]. Because of uncertainties involved with radiological detection at extremely low levels, ATSDR health physics staff use a value of 3 to 5 times greater than the suspected background values as an indicator of significance. Therefore, the reporting level of 0.005 pCi/g (2×10^{-4} Bq/g), a value twice the reported background, is considered valid for the purposes of the sampling plan.

Analytical processes report a minimum detectable activity or level (MDA or MDL). Table I summarizes the numbers of samples, the number of samples in which the MDA value was above the reporting level and the number of samples in which the detected activity was below the MDA. Because the MDA is a variable that depends on the sample characteristics and instrument operational restraints (background, counting time, etc.), the detector instrumentation and their computer related programs can report values below the MDA once these restraints are taken into account. The computer-estimated values below the MDA are usually reported as either the MDA or non-detects; that is, no activity in the sample under investigation.

[3] The geometric mean is used to measure the central tendency of a data set, especially if the data set is not evenly distributed.

When all samples collected from Big Trees Park are reviewed as an entire group, the following information is observed. The highest concentrations of the radionuclides and their locations were as follows: A) Pu 238 detected at $8.57 \times 10^{-2} \pm 0.018$ pCi/g (3.17×10^{-3} Bq/g) in grid location G0103 (10 cm depth); B) Pu 239/240 at 0.774 ± 0.11 pCi/g (2.9×10^{-2} Bq/g) in grid location G0103 (10 cm depth) and; C) Am 241 at 0.205 ± 0.066 pCi/g (7.6×10^{-3} Bq/g) in grid location G0104 (5 cm depth). For comparison, the EPA Region IX Preliminary Remediation Goal is 2.5 pCi/g for Pu 239/240 and the National Council on Radiation Protection and Measurements (NCRP) recommends approximately 51 pCi/g for each of these radionuclides in a park scenario to be used as screening level indicator for the evaluation of additional actions [10]. The most stringent NCRP value is 7.8 pCi/g for a sparsely vegetated rural area.

Table I. Data Summary for Big Trees Park 1998 Sampling Effort

Radionuclide (reporting level)	Total number of samples[1]	Number above reporting level	Number below MDA (percent)
Am 241[2]	117	12	105 (90)
Pu 238[3] (0.005 pCi/g)	379	34	287 (76)
Pu 239/240[3] (0.005 pCi/g)	379	94	213 (56)

1. Numbers include field replicates on approximately 10% of the samples initially collected.
2. Analysis performed by low energy gamma spectroscopy and alpha spectroscopy.
3. Analysis performed by alpha spectroscopy that is, specific for the listed radionuclide.

Pathway Analyses - Radiological Considerations

Air pathway The soils samples used to evaluate the air pathway were collected from the grid locations (coded GXXX where XXX represents numbers). Collection depths ranged from 5 cm to a depth of 40 cm. The analysis of these samples was for Pu 238, Pu 239/240, and Am 241. Only those samples that met the criteria previously discussed were used in the analysis; in all 111 samples from all depths met ATSDR's pre-established criteria.

Several issues are to be considered for evaluating this pathway. Among these are: 1) the size and location of the grid; that is, is there sufficient separation of the sampling locations to give adequate spatial distribution of samples; 2) the comparison of the grid with other locations within the park, and; 3) comparison of the grid locations with other Livermore Valley sampling locations considered background and listed in the sampling plan [2].

The air pathway analysis was performed by comparing the Pu concentration in the grid area using the top 5 cm samples and comparing those to the Pu distribution at 5 cm depths from other locations in the park, comparing the 5 cm depths to the 10 cm and 20 cm depths, and from 5

other locations near the laboratory and south of the laboratory as listed in the sampling plan [2]. These samples, listed in the sampling plan, are coded HOSP, MET, MESQ, ERCH, and RRCH. For the analysis, ATSDR used analysis tools for investigating small numbers of samples called the t-test and the F-test [11]. A t-test evaluates the average or mean of a distribution of samples to determine if they are similar. One result of this test is the calculation of a "t value" that can be compared to a tabulated value. The tabulated value is called the critical value, found in any book on statistics, is used in business, scientific, or any other discipline in which a statistical analysis is required. If this "t value" is less than the tabulated value, then there is no difference between the values evaluated. In a similar manner, the F-test is a procedure to determine if the variation in the sample measurements is significant. If the measured "f value" is greater than the critical tabulated value, then the chance that the samples are identical is small [11]. The grid sample values also were entered into a geographical information system (GIS) to display the spatial configurations to determine if a distribution pattern could be visualized.

If the Pu reached the park via the air pathway, the Pu concentrations in the associated grid locations should be significantly different from the Pu at other surface areas in the park. This would not necessarily rule out the air dispersion pathway as other locations within the park could have been impacted by the air releases. If the Pu in the top 5 cm of soils is significantly different from that in the samples collected 10 and 20 cm, then either air dispersion or other methods of distribution are possible. The other 5 sampling locations listed in the sampling plan were chosen for the following reasons. First, the winds in the Livermore valley, based on wind direction reports show that little wind, if any come from the north; therefore the ERCH samples could represent a background location [7]. The HOSP samples are west of the ERCH samples and could also be considered similar to the background samples. The MET and MESQ samples are at the west boundary of LLNL, and these samples could be expected to be impacted by air releases when the wind blows from easterly to westerly directions. The RRCH location, north of Interstate 580 near Vasco Road, is a downwind direction when the wind blows from the south across the laboratory property.

The results of the analyses of these data are given in Table II. In essence, the soils in the top 5 cm from the grid locations are not significantly different from the soils collected from the other 5 cm samples from the areas within the park, from the soil samples collected at 10 cm in the grid area, or from soils collected in the top 45 cm of the tree wells. At other areas, both in the park and outside the park, the 5cm depth samples were significantly different. Those samples at the laboratory boundary, MET and MESQ, have Pu concentrations lower than soil samples collected in the park. This would not be expected if air dispersion were the pathway- Pu is a dense material and would deposit at higher concentrations closer to the release point. Furthermore, the grid samples are essentially identical across the park and in the tree wells which apparently received the sewerage sludge; this type of distribution would not be expected if the Pu had been deposited via the air pathway.

In those areas that are downwind of the laboratory, the concentrations of Pu in soils are higher than those concentrations found in the park. In 1980, the California Department of Health

Services collected soil samples from areas east, northeast, and east northeast at a distance of up to 2 kilometers (1.2 miles) from LLNL, approximately the same distance Big Trees Park is from LLNL. These directions are downwind of LLNL the majority of the time [12]. The geometric mean concentration of Pu 239/240 from these locations was 0.068 pCi/g ($2.5x10^{-3}$ Bq/g) as compared to the geometric mean of 0.017 pCi/g (6.3×10^{-4} Bq/g) for the top 5 cm throughout the park. This would suggest that Pu in the park is not the result of atmospheric deposition from LLNL activities.

The GIS spatial analysis indicated that the Pu is not uniformly distributed and these values did not fit a continuous distribution as would be expected from an atmospheric dispersion pattern.

Table II. Statistical Comparison of Big Trees Park Grid Locations to Other Areas

Location	t-test value[1]	critical T value[2]	X[7]	F-test value[3]	critical F value[4]	X[7]
Other park locations[5]	1.9	2.03	X	46	8.6	
Tree Well[6]	1.1	2.03	X	35	2.7	
Outside Tree Well[6]	2.8	2.04		>100	2.9	
Grid (10 cm)	-0.5	2.02	X	0.56	0.54	
Grid (20 cm)	2.14	2.03		20	2.0	
HOSP	2.7	2.04		>100	3.1	
MESQ	2.9	2.04		>100	3.4	
MET	2.9	2.04		>100	3.4	
ERCH	2.8	2.04		>100	4.5	
RRCH	2.8	2.04		>100	3.8	

1. The t value represents the value that defines how the means (averages) differ. A negative t value indicates the grid average was lower than the other areas to which it was compared.
2. The critical T value is the 95% confidence interval in which the t-value must fall for the samples to be considered identical.
3. The f value represents the value that defines the ratio of the variances.
4. The critical F value, the maximum value, which indicates that the ratio of variances is caused by chance alone.
5. Comparison of other soils samples in the park at a depth of 5 centimeters.
6. Comparison of soils collected from 0 to 45 centimeters.
7. Indicates sample is not significantly different at the 95% confidence level.

Is the Pu in the park the result of atmospheric fallout? The question has been raised as to the chance that the Pu in Big Trees Park is the result of atmospheric nuclear testing. All nuclear

weapons detonated in the atmosphere release various amounts of Pu radioisotopes at different isotopic ratios. Over time, these ratios vary as the material decays. An analysis of the Pu and Am found in the park can be compared to these measured fallout levels. The ratios of the values in the park and those found in fallout is given in Table III. Based on this analysis, the Pu in the park does not appear to be the result of atmospheric fallout as the ratios are 4 to 15 times higher than values normally associated with fallout deposition.

Table III. Isotopic ratio of radionuclides in Big Trees Park and in fallout.

Ratio	Big Trees Park (all samples)[1]	Fallout[2]
Am 241/Pu 239+240	1.22	0.33
Pu 238/Pu 239+240	0.53	0.065 (0.036 in Northern Hemisphere)

1. Samples that met ATSDR requirements for acceptable samples.
2. Data from Efurd, D.W., et al., (1997). Evaluation of the anthropogenic radionuclide concentrations in sediments and fauna collected in the Beaufort Sea and Northern Alaska. Los Alamos National Laboratory. LA-13302-MS, UC-000 and UC-702 [13].

Conclusion: Based on the analyses of the grid samples and comparing them to areas where air deposition is known to have occurred, ATSDR does not believe that the presence of plutonium in Big Trees Park is the result of air deposition.

Arroyo Seco Pathway Samples collected to test the hypothesis that Pu was released to the former Arroyo Seco channel are coded beginning with B-FAS. The hypothetical pathways for Pu entering the arroyo would include bulk flow (overland flow) from the southeast corner of the laboratory or via a possible sewer line rupture. If Pu was released to the arroyo, samples collected in the former arroyo channel or the current channel would indicate this as the pathway whereby Pu reached the park. The results of the samples from the previous arroyo channel were below the MDA, the highest computer-estimated value was 0.00345 pCi/g (1.3×10^{-4} Bq/g) for Pu 238 and 0.00229 pCi/g (8×10^{-5} Bq/g) for Pu 239/240. No Am 241 was detected.

For the samples collected in the current Arroyo Seco (coded SSS-AS) duplicate samples were collected at each location. The highest Pu 238 computer estimated value detected was less than the MDA; that value was 0.00269 pCi/g (1×10^{-4} Bq/g). For Pu 239/240, of the two samples collected at a depth of 25 centimeters below the surface downstream from the concrete channel, one sample was identified at a concentration of 0.0432 pCi/g (1.6×10^{-3} Bq/g) and the duplicate was below the MDA. Pu 239/240 was only detected in one other sample at this depth, that sample location also was downstream of the concrete channel. The duplicate for that sample was below the MDA as well. Samples obtained from the arroyo immediately downstream from the LLNL outfall near East Avenue and Building 111 on LLNL property, and upstream of the outfall showed no Pu activity; that is, all were below the MDA. Furthermore, all samples collected a depth of 5 centimeters were below the reporting level or less than the MDA.

Conclusion: Based on the results, the current Arroyo Seco channel does not appear to be the source of Pu in Big Trees Park. Although 2 samples suggest the presence of Pu, the duplicate samples do not validate these apparently elevated levels. This suggests either a random deposition of Pu from areas currently in the park, a random error associated with Pu deposition, or a possible error in the laboratory analysis. Nonetheless, because no other samples collected in the arroyo at either 5 cm or 25 cm contained Pu above the detection level, there is no indication that Pu ever entered the Arroyo Seco from the LLNL outfall in the southwest corner of the site, from overland transport from the southeast corner of the facility to surface collection areas that enter the outfall areas near Building 111, nor from any reported sewer line break.

Sewage Sludge Pathway The investigation of this pathway was through analysis of samples collected around the ornamental trees. These samples, coded TR, were collected around 10 trees and at depth intervals of 0 to 45 cm, 45 to 90 cm, and 90 to 135 cm. The soil samples were taken inside the tree well and outside the tree well. The tree well is the hole dug prior to planting. If soil amendments containing sewage sludge were added during the planting, the radionuclide concentrations inside the well compared with the soils from similar depth intervals outside the wells could be used for these analyses. For samples to be valid for analysis, 2 criteria had to be met. Firstly, the detected concentration had to be greater than the MDA; secondly, the detected concentration had to be greater than the 2σ error of the measurement. Tree well samples and samples from outside the tree well that do not meet these criteria can be considered indistinguishable.

Upon analysis, 8 ornamental trees fit the criteria for one or more of the radionuclides under consideration. It is interesting to note that only the top 0 to 45 cm sample from each of the trees suggested the presence of elevated concentrations of radionuclides. The ratios associated with these trees are shown in Table IV. For those Pu 239/240 samples within the tree wells, the 2σ error average was less than 25% indicating the detected concentration was significantly higher than the MDA. Conversely, outside the tree well, the 2σ error averaged 67% suggestive that these samples are more closely associated with nominal background concentrations.

Conclusions: From this analysis, the following information can be deduced: 1) The ratio of radionuclides at depths greater than 45 cm within the tree well as compared to the outside area are not significantly different from background; 2) Pu or Am radionuclides do not increase with depth; 3) Because the concentrations of these radionuclides at depth (greater than 45 cm) are indistinguishable from background, no widespread distribution of radioactive materials has occurred and; 4) Sewage sludge containing Pu radionuclides was most likely applied to these ornamental trees as the area outside the tree wells is not contaminated.

Table IV. Ornamental trees with elevated ratios of radionuclides

Tree	Pu 238 ratio	Pu 239/240 ratio	Am 241 ratio
2	NS [1]	NS	NS
3	NS	NS	0.067
4	NS	0.84 [2]	NS
5	NS	20.7	NS
6	3.6	23.4	NS
8	NS	19.4	NS
11	NS	12.6	NS
12	NS	14.9	NS

1. NS - indicates that the criteria for evaluation were not met; therefore, these ratios are not considered significantly different.
2. For this tree, the criteria were met for evaluation but the ratio was not elevated.

Evaluation of Other Sampling Locations

Eastern Extension of Big Trees Park Soil samples collected from the eastern extension of Big Trees Park (coded BPE) did not contain any Pu 238 above the reporting limit. One sample obtained from a depth of 40 cm was above the MDA with a reported concentration of 0.0019 pCi/g (7 x 10^{-5} Bq/g). Twenty-five soil samples were analyzed for Pu 239/240; all values were below the reporting level but 8 samples were above the MDA. Of the 8 samples, duplicate samples were collected in 7 instances. The duplicates were below the MDA. Table V gives the synopsis of the information for these 8 samples.

Disced Area Four samples (coded DISK) collected in this area were analyzed for Pu radionuclides. The results of the Pu 238 analysis indicate that no Pu 238 was found above the detection limit. The analysis of samples for Pu 239/240 showed that all samples were below the reporting limit; however, one sample was above the MDA. That sample has an associated error of approximately 75% and is questionable with respect to its reported value.

Playground Pu 238 was not detected in soil samples from the playing field (coded PLAY) above the reporting value of 0.005 pCi/g (2 x 10^{-4} Bq/g) with most samples being reported at less than the MDA. The few samples in which the value was greater than the MDA also were suspect as the associated error of the measurement was more than 70%. Fifteen soil samples were collected for Pu 239/240 analysis with one sample from a depth of 40 cm exceeding the reporting limit and one sample from 5 cm essentially at the reporting limit. Sample duplicates did not contain Pu 239/240; that is, the reported values were below the detection limit.

Table V. Sampling: results from the Big Trees Park Eastern Extension[1]

Sample Location (depth in centimeters)	Pu -239/240 (picocuries per gram)	Minimum Detectable Activity (picocuries per gram)	Reporting Limit (picocuries per gram)
L-BPE01 (5)	*0.000996 ± 0.00165* 0.0399 ± 0.00958	0.00282 0.00126	0.005
L-BPE01 (20)	-0.000469 ± 0.00094 0.0121 ±0.00478	0.00311 0.00318	0.005
L-BPE02 (5)	0.0429 ± 0.00895 *0.00175 ± 0.00145*	0.0025 0.000877	0.005
L-BPE02 (10)	*0.000308 ± 0.000617* 0.00467 ± 0.00251	0.000924 0.00225	0.005
L-BPE02 (20)	0.00395 ± 0.00259 *0.000655 ± 0.00124*	0.00271 0.00236	0.005
L-BPE02 (30)	*0.00492 ± 0.00253* 0.000675 ± 0.000958	0.000922 0.00101	0.005
L-BPE03 (10)	0.00416 ± 0.00291	0.00367	0.005

1. Values in small, italics type represent duplicate samples that are not above the MDA.

Pathway Analyses - Metals Considerations

Samples for metals analysis were collected from the ornamental trees area and the grid area. With respect to samples collected from the ornamental tree area, sample collection was from inside and outside the tree well. All metals were detected above the MDA. ATSDR believes that if sludge had been used as a soil amendment, the amendment would be used either as a mulch-type material applied to the surface around the tree trunk or uniformly mixed in with soil in which the tree was planted – the tree well. Therefore, the ratio of individual metals at each depth compared with the same metal concentrations outside the tree well area at the same depths should answer this question.

Sampling was around 10 trees and at depths of 0 to 45 cm, 45 to 90 cm, and 90 to 135 cm. To evaluate the results, a ratio was calculated by dividing the concentration of the individual metals in the tree well at a specific depth by the concentration of the same metals at the same depth outside the tree well. If the concentration inside the tree well is higher than the concentration outside the tree well, the ratio is greater than 1.0; the higher the ratio, the higher the concentration inside the tree well. If the ratio is greater than 2.0, ATSDR considered the concentration inside the tree well to be significantly different that the concentration outside the tree well. Many trees did not show significant differences in metals concentrations at any depth between the tree well and

outside the tree well. Results from Tree 3, the tree with the greatest differences, are given in Table VI. Only the concentrations of the metals in the top 45 cm of soil in the tree well were elevated; the samples at the deeper depths are not elevated. This is reflected in the ratio of metals concentrations as shown in Table V as well. **These data suggest that sewage sludge was added to the surface soils, at least around this tree, as the concentrations in the top 45 cm is about 8.5 to 10 times that at found outside the tree wells.**

Table VI. Ratio of metals concentration associated with Ornamental Tree 3[1]

Depth (cm)	Chromium	Copper	Lead	Nickel	Zinc
0-45	9.89	8.95	8.45	9.62	6.85
45-90	1.25	1.57	1.44	1.38	1.47
90-135	0.94	0.95	0.92	0.96	1.01

1. The ratio is the concentration inside the tree well as compared to outside the tree well.

Metals analysis of samples collected from the grid area suggests that the concentrations of metals in this area are similar to the metals concentration in those samples collected outside the tree wells. Soil samples from the grid that were collected from depths ranging from 5 to 40 cm exhibited the same concentrations as soil samples collected at a minimum depth of 0 to 45 cm from outside the tree wells. Conversely, the metals concentrations in grid samples were lower than the metals concentration of the soil sample collected from within the tree wells. **This also suggests that sewage sludge was added as a soil amendment to the trees.**

Health impacts of heavy metals in Big Trees Park

ATSDR has established environmental media evaluation guides (EMEG) for many common heavy metals found in the environment. EMEGs represent the concentration in an environmental media below which no adverse health effects are expected to occur. For the heavy metals reported in Big Trees Park, only the only EMEG established is for zinc with an EMEG of 600 milligrams per kilogram (parts per million, ppm). The highest concentration of zinc reported was about 74 ppm. Another evaluation guide used by ATSDR, the reference media evaluation guide or RMEG is listed for nickel. The RMEG for nickel is 40 ppm for chronic ingestion by a pica child. Although 40 soil samples from Big Trees Park exceeded this RMEG, only 2 samples collected from the grid were at the surface (5 cm depth). However, throughout the entire park, the average nickel concentration was less than this RMEG.

Conclusion: ATSDR does not believe any adverse health effects from nickel would occur based on site wide concentrations and depth of contamination.

Is there a need for additional sampling in the Livermore Valley? As previously discussed [1], sewage sludge containing Pu released from LLNL was distributed on demand to the community. The log book indicating who received this sludge has not been available [1]. Nonetheless, LLNL in May 1973 sampled the yards of three laboratory individuals who received the sludge, knew where the sludge had been placed, and knew what types of soil disturbing activities had taken place since the addition of the sludge to those yards. Table VII shows the results of the 1973 sampling.

Table VII. Results of 1973 sampling; in yards receiving sewage sludge[1]

Location and depth	Pu 238 ± 2 sigma (%)[2]	Pu239/240 ± 2 sigma (%)[2]
#1, 0 to 1 cm	0.0233 ± 10	0.324 ± 9.8
#1, 1 to 25 cm	0.00023 ± 11.6	0.00412 ± 7.4
#2, 0 to 1 cm	0.243 ± 13	1.84 ± 13
#2, 1 to 25 cm	0.0964 ± 9.1	0.784 ± 9
#3, 0 to 1 cm	0.000923 ± 19	0.00797 ± 8.8
#3, 1 to 25 cm	0.00013 ± 46	0.00168 ± 15

1. Concentrations expressed as pCi/g.
2. The error is expressed as a percentage. For example 10 ± 1 is expressed as 10 ±10%

At Location 2 where the highest concentration of Pu 239/240 was detected, the concentration is below the EPA Preliminary Remediation Goal of 2.5 pCi/g and below the NCRP recommendation for additional action of 51 pCi/g [10]. Therefore, these yards would not be considered a candidate for any additional activities and no additional sampling would be warranted.

ATSDR bases this conclusion on the following. The contamination in excess of nominal background concentrations occurred in the top 1 centimeter (top 3/8 inches). In the 25 or more years since the sludge had been distributed, any one or more of the following events could have occurred: 1) exterior home remodeling; 2) extensive landscaping; 3) normal weathering such as erosion of soils and general yard maintenance activities; 4) new owners who would not know if sludge had been used and; 5) city expansion into outlying areas or purchase of pre-existing private land. Because of these possible activities, any sampling locations would be very difficult to identify and, if Big Trees Park and the employees' yards are a model, then the Pu possibly detected would be below federal action levels.

CONCLUSIONS

An extensive sampling plan and sampling activities were implemented to evaluate potential pathways whereby Pu reached the Big Trees Park in Livermore, California. The pathways evaluated included air dispersion, dredging of the Arroyo Seco, sewage sludge distribution, and atmospheric fallout deposition from LLNL activities.

Based on this analysis, ATSDR believes the most credible pathway whereby Pu radioisotopes were introduced into Big Trees Park was the application of sewage sludge as a soil amendment. This is based on a comparison of the concentrations of both Pu and heavy metals collected from within tree wells to the concentrations present outside the tree wells. Although Pu is elevated in the park, these levels are below levels of both health concern per EPA Region IX Preliminary Remediation Goals and below the recommended levels requiring additional activities as discussed by the NCRP. No other pathway appears to be a viable pathway for its presence.

RECOMMENDATIONS

No further actions are recommended.

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